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Potentially toxic elements (PTEs) pollution in surface soils in a typical urban region of south India: An application of health risk assessment and distribution pattern

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Highlights

- Potentially toxic elements (PTEs) contamination levels were estimated by using profound methods such as contamination factor, degree of contamination and index of geo-accumulation.
- Assessment of non-carcinogenic and carcinogenic risks for children and adults were investigated in the study region.
- Principal component analysis of potentially toxic elements were studied and also generated their spatial distribution maps in the investigated region.

Abstract:

The pollution level of potentially toxic elements (PTEs) in surface soils is detrimental to the ecosystem and human health. In this research, various indices such as an index of geo-accumulation (I_{geo}), contamination factor (*CF*), degree of contamination (*DC*), and principal component analysis (PCA) were implemented to identify and evaluate the soil PTEs pollution; and then human health risk assessment model used to establish the link between heavy metals pollution and human health in the urban region of south India. Results exhibited that the mean concentration of Cr, Cu, Ni and Zn were found to be 1.45-6.03 times greater than the geochemical background values. Cr and Cu were the most profuse PTEs measured in the soils. The pollution indices suggest that soil of the study region is mainly moderate to highly polluted. The non-carcinogenic health risk assessment proposed by the United States Environmental Protection Agency (USEPA) suggested the mean hazard indices (HIs) were below one which denotes no significant of non-carcinogenic risks to both children and adults. Furthermore, carcinogenic risk assessment results advised ~80% of cancer risk was caused by Cr contents, while other heavy metals indicate that neither children nor adults in the study region were of carcinogenic risks.

Keywords: Surface soils; potentially toxic elements; Pollution characteristics; Health risks; South India

1. Introduction

Due to the rapid development of urbanization and continuous growth of the industrial segments, the severe pollution of soils by increasing the concentration of potentially toxic elements (PTEs) which has greatly caused widespread concern in many developing countries, due to PTEs are typically harmful to the environment and also endanger to human health (Adimalla, 2020b; Adimalla et al., 2020; Baltas et al., 2020; Jiang et al., 2020). Therefore, in recent years most of the researchers/scientists focus on PTEs pollution in soils, contamination process, and source identification by using various geostatistical methods and also its concomitant human health risks in various regions in the world. For example, Baltas et al. (2020) have studied the PTEs (Cr, Fe, Ni, Cu, Zn, As and Pb) pollution in agricultural soils around Sinop province, Turkey, and found the mean concentrations of PTEs (Cr, Ni, As, and Pb) surpassed their threshold level due to the Sinop region was greatly influenced by anthropogenic inputs. Additionally, they also evaluated the health risks, their results indicated that the children were effectively influenced by the non-carcinogenic and carcinogenic health risks of PTEs (Baltas et al., 2020). Jiang et al. (2020) focused on the sources of soil PTEs pollution by using an integrating geostatistical method in the Guangdong region of southeastern China. Their results displayed the mean concentrations of zinc, lead, arsenic, mercury and cadmium in soil were exceeded the corresponding background values. Furthermore, they also noticed four possible contamination sources in Guangdong region soils such as industrial activities, agricultural practices, natural source and traffic emissions (Jiang et al., 2020). Cicchella et al. (2020) emphasized on the urban soil contamination in the city of Salerno, Italy, and they observed that the Salerno urban soils were affected by moderate to high contamination and extensively within highly populated areas, industrial sites and also along high traffic roads. In addition, they also noticed that most of the heavy metal concentration values in the Salerno area soils were an order of magnitude and higher than their background values which strongly indication of a direct correction with anthropogenic sources. Therefore, the above comprehensive study profoundly divulges the PTEs typically endanger to human health because of their non-biodegradability, toxicity and persistence (Adimalla, 2020a; Baltas et al., 2020; Konstantinova et al., 2019; Sun et al., 2019; Zhao et al., 2019). Specifically, lower concentration of PTEs like Ni, Mn, Fe, Zn and Cu are recognized as micronutrients which are mostly regulating the physiological function of the human body (Chakraborty et al., 2019; Giri et al., 2017; Jiang et al., 2019; Zhuo et al., 2019). Conversely, a few PTEs are like Cr, Pb, Cd and As have typically no recognized physiological risks on humans but they can show toxicity/health-risks even at low concentrations (Adimalla, 2020b; Adimalla and Wang, 2018; Deng et al., 2019; Kaur et al., 2019). In-depth research has profoundly documented that continuous exposure to PTEs can cause many negative effects on human health such as mental retardation, a verity of cancer, cardiovascular, kidney and also neurological diseases.

Soil PTEs pollution has also been a widespread environmental problem in India for the last few decades (Adimalla et al., 2020; Adimalla et al., 2019; Kashyap et al., 2019; Kumar et al., 2019; Naz et al., 2018). Many researchers like Kashyap et al. (2019); Adimalla 2020a, b; Kaur et al. (2019); Kumar et al. (2019); Giri et al. (2017); Adimalla and Wang (2018); Adimalla et al. (2019); have literally studied the PTEs contamination in soils of various regions in India. However, the present investigation region falls in the part of the Sangareddy district of Telangana state, India which is the most intensively developing urban region. Importantly, in the last few years, the urban population has doubled, and the urban area and transportation system have significantly developed. However, to the best of our knowledge, no studies had been carried out on the comprehensive evaluation of spatial distribution characteristics of soil PTEs and its associated human health risks posed by PTEs in surface soils in the examined region. Therefore, to reduce the gap, the main objectives of our present investigation were to (1) determine the concentration of the PTEs and also evaluate the spatial distribution mapping to get a clear visual picture of PTEs, (2) analyze the degree of soil contamination by using geo-accumulation index (I_{geo}), contamination factor (CF) and degree of contamination (*DC*), and (3) ascertain the possible potential risk of local residents (children and men). The outcome of this study can surely provide scientific base-line information for which to estimate future soil quality measures in the investigation region.

2. Materials and methods

2.1 Study region

The present examined region is situated on the western part of the Sangareddy City and lies between longitudes 77.50º to 77.67º E and latitudes 17.75º to 17.83º N covering an estimated area of 125 Km^2. The area has a population of about 1,527,628 people based on the 2011 census of India (Census 2011) and an average population density of 340 people/Km^2. Typically, the study region is considered by the distinct dry and wet season, with an average annual rainfall of the district is 910 mm, while the mean temperature in the range of 13-38.8ºC. The geological formations of the study region are well documented (Adimalla, 2020a; Adimalla and Taloor, 2020; Adimalla and Venkatayogi, 2017; Dantu, 2014). The geological formations in the study region are predominantly dominated by basalts and laterites which are obviously depicted in Fig 1. The major part of the study region is covered by laterites. These laterites majorly ensue as cap rocks over the basalts with an elevation ranges from 600 to 660 mean sea level (MSL). Furthermore, in the study region, basalts mostly display both vesicular and non-vesicular texture. The majority of the study region soil is covered by black and reddish-brown in color.

2.2 Field Sampling

A total of twenty composite soil samples (0-10 cm depth) were collected for the present study region, and each sampling location (ZSI-1 to ZSI-20) was recorded by using a portable global positioning system (GPS: Garman eTrex 30). Figure 1 unveils the location map of the investigated region and with soil sampling locations. Especially, each composite soil sample consisted of five sub-samples from randomly selected positions around the sampling site. Finally, each soil sample was placed in properly labeled polythene bags and transported to the laboratory for analyses.

2.3 Sample analysis

The collected soil samples were scrupulously air-dried for 48 h to 60 h. These dried samples were then disaggregated with mortar and pestle. Finally sieved through -200 mesh size (US Standards) using a swing-grinding mill. Boric acid is used to prepare sample pellets by applying pressure at 25 tones (Herzog make) for XRF analysis to determine heavy metals. Aluminim cups are used to prepare the pellets. A fully automated Philips MagiXPRO-PW2440, microprocessor-controlled, 168-position automatic PW-2540 vrc sample changer wavelength dispersive X-ray spectrometer is used along with 4KW X-ray generator for the determination of heavy metals in the soil samples. International soil reference materials were used to prepare calibration curves for different potentially toxic elements and to check the accuracy of the analytical data. Canadian soil reference materials SO-1 and SO-4 were used to estimate the analytical bias of the data of the soil samples and details are listed in Supplementary Table S1. It can be seen from Table S1, the present study analytical values were found to be within the certified values of the standard soil reference materials which confirms the reliability of the PTEs analysis results.

2.4 Contamination factor (*CF*)

In the early 1980s, the Hakanson has developed a profound mathematical model to evaluate the degree of soil contamination by heavy metals (Hakanson, 1980). *CF* is calculated using the following equation:

$$CF = \left({C_{0-1}^{i}} \middle/ {B_n^i} \right) \qquad (2)$$

Where C^i_{0-1} refers to an average concentration of PTEs of at least five sampling sites and B^i_n is the concentration of the same toxic elements of soils in Medak (Dantu 2014). To assess the degree of contamination of PTEs, Hakanson (1980) categorized the *CF* into four classes such as $CF<1$: low contamination, $1\leq CF\leq 3$: moderate contamination, $3\leq CF\leq 6$: considerable contamination and $CF>6$: very high contamination (Hakanson, 1980).

2.5 Degree of contamination (*DC*)

The degree of contamination (*DC*) is widely used to characterize and estimate the contamination of soil PTEs which is proposed by Hakanson (1980). Fundamentally, the degree of contamination, i.e. the sum of all contamination factors (*CF*) for a given soil heavy metals. *DC* is computed using the following equation.

$$DC = \sum_{i=1}^{m} CF \qquad (3)$$

Where *CF* is the contamination factor and "*m*" the count of metals species. For evaluating the degree of contamination, four categories have been suggested by Hakanson (1980): $DC<8$: low degree contamination, $8 \leq DC < 16$: moderate degree of contamination, $16 \leq DC < 32$: considerable degree of contamination and $DC > 32$: very high degree of contamination.

2.6 Index of geo-accumulation (I_{geo})

Mueller introduced a technique/method called "Index of geo-accumulation (I_{geo})" in the year 1969. This method enables us to measure the anthropogenic influence of PTEs contamination in media that include soils, dust, and sediments in aqueous environments (Adimalla, 2020b; Adimalla et al., 2020; Baltas et al., 2020; Jiang et al., 2019; Muller, 1969). The I_{geo} is calculated using the following equation:

$$I_{geo} = log_2\left(C_n^{HMs} \middle/ 1.5 \times B_n \right) \qquad (4)$$

Where C_n^{HMs} refers to the measured concentration of PTE "*n*" (mg/kg), and B_n represents the geochemical background value for the PTE "*n*" (mg/kg). In this study, B_n values were taken from Dantu (2014) for the calculation of I_{geo} and *CF*. The constant factor 1.5 is introduced to reduce the effect of possible variations in the B_n values that are due to lithologic variations in the surface soils. The I_{geo} scheme is classified into seven subclasses like Class-0 ($I_{geo} \leq 0$ uncontaminated), Class-1 ($0 < I_{geo} \leq 1$ uncontaminated to moderately contaminated), Class-2 ($1 < I_{geo} \leq 2$ moderately contaminated), Class-3 ($2 < I_{geo} \leq 3$ moderately to heavily contaminated), Class-4 ($3 < I_{geo} \leq 4$ heavily contaminated), Class-5 ($4 < I_{geo} \leq 5$ heavily to extremely contaminated) and Class-6 ($I_{geo} > 5$ extremely contaminated) (Muller, 1969).

2.7 Human exposure and health risk assessment model

The health risk assessment model was initially proposed by the United States Environmental Protection Agency (USEPA) appraise and envisage the possible deleterious effect on human health due to perpetual exposure of toxic elements by various exposure pathways (USEPA, 1989, 1997). This profound model enables us to evaluate both non-carcinogenic and carcinogenic risk by three potential exposure pathways including oral ingestion, inhalation via nose, mouth, and dermal contacts (USEPA, 1989, 1997).

2.7.1 Non-carcinogenic risk

Typically, the non-carcinogenic health risk from PTEs is articulated by the hazard quotient (HQ_i). The HQ_i is assessed by average daily exposure dose (ADD) of each PTE and its corresponding reference dose (R*f*D). Finally, the non-carcinogenic health risk is computed by using the following equations:

$$ADD_{ing} = \frac{C_{soil} \times IngR \times EF \times ED}{BW_A \times ET_A} \times 10^{-6} \tag{5}$$

$$ADD_{derm} = \frac{C_{soil} \times ESA_s \times AF_s \times EF \times ED}{BW_A \times ET_A} \times 10^{-6} \tag{6}$$

$$ADD_{inh} = \frac{C_{soil} \times InhR \times EF \times ED}{BW_A \times ET_A \times EF_p} \tag{7}$$

$$HI = \sum HQ_i = \sum \frac{ADD_i}{RfD_i} \tag{8}$$

Where ADD_{ing} means the average daily exposure dose through ingestion pathway (mg/kg/day), ADD_{derm} is the average daily exposure dose through dermal contact pathways (mg/kg/day), ADD_{inh} represents average daily exposure dose to particulate in soils through inhalation pathway (mg/kg/day), C_{soil} is the concentration of PTEs in soil (mg/kg). $IngR$ and $InhR$ are the ingestion (mg/day) and inhalation rates (m^3/day) of the soil particles, respectively. EF is the exposure frequency (day/year), ED is the exposure duration (year), BW_A is the average body weight of exposed individual (kg), ET_A is the average exposed time (days), AF_s is the skin adherence factor (mg/cm^2), ESA_S is the exposed dermal skin surface area (cm^2), *RfD* is the reference doses, EF_p is the particle emission factor (m^3/kg). *HI* is the total non-carcinogenic health risk posed by exposure of multiple exposure pathways. If *HI* is smaller than one, the non-carcinogenic health risk is relatively overlooked while *HI* is larger than one, the non-carcinogenic health risk is significant.

2.7.2 Carcinogenic risk

Typically, carcinogenic risk (*CR*) reveals the possibility of the development of cancer risk due to the various exposure pathways (USEPA, 1989). The individual carcinogenic risk (*CR*) and total carcinogenic risk (*TCR*) are basically estimated by using the following equations:

$$CR_{ing} = \frac{C_{soil} \times IngR \times EF \times ED}{BW_A \times ET_{ca}} \times 10^{-6} \times SF_{ingestion} \qquad (9)$$

$$CR_{derm} = \frac{C_{soil} \times ESA_s \times AF_s \times EF \times ED}{BW_A \times ET_{ca}} \times 10^{-6} \times SF_{dermal} \qquad (10)$$

$$CR_{inh} = \frac{C_{soil} \times InhR \times EF \times ED}{BW_A \times ET_{ca} \times EF_p} \times SF_{inhalation} \qquad (11)$$

$$TCR = \sum (CR_{ing} + CR_{derm} + CR_{inh}) \qquad (12)$$

Where CR_{ing}, $CR_{derm,}$ and CR_{inh} represent the ingestion, dermal, and inhalation pathways of *CR*, and *SF* is the carcinogenic slop factor of PTEs (mg/kg/day). ET_{ca} and *SF* are the carcinogenic average exposed time (days), and slope factor (mg/kg/day), respectively. There is no significant health risk when the values of *TCR* are in the range of 1×10^{-6} to 1×10^{-4}. However, it exceeds the limit causes serious health hazards. Definitions and reference values of both non-carcinogenic and carcinogenic risks presented in equations 5 to 12 are clearly recorded in Table S2 as obtained from the relevant literature. According to USEPA database, R*f*D and *SF* values in various exposure pathways are listed in Table S3.

3. Results and discussion

3.1 Descriptive statistics

Table 1 divulges the descriptive statistics (minimum, maximum, mean, standard deviation, coefficient of variation, skewness, and kurtosis) of six PTEs in the soils from the study region. The concentrations of As, Cr, Cu, Ni, Pb, and Zn in soils varied from 2.3 to 4.8 mg/kg, 158 to 482 mg/kg, 84 to 214 mg/kg, 19 to 51 mg/kg, 3.1 to 32 mg/kg and 84 to 134 mg/kg, respectively. The mean concentrations of Cr, Cu, Ni, and Zn were 6.03, 3.45, 1.64, and 1.45 times larger than their corresponding geochemical background values, respectively. Furthermore, Table 1 also discloses that the mean concentrations of As and Pb did not exceed their corresponding geochemical background values in the study region soils. However, large standard deviations were noticed in all studied PTEs except As (Table 1), suggesting the wide variation of concentrations in soil samples in the study region. Skewness values of Cr and Cu are larger than 1, demonstrating these two PTEs positively skew towards lower concentrations. This can be confirmed by the median concentrations of Cr and Cu are considerably smaller than their mean concentrations. As a result, the K-S test confirmed for these two PTEs in the investigated region soils were only recorded as bigger than 0.2 which means these PTEs were normally distributed (Table 1).

In general, the coefficient of variation signifies the various dimensions of the indicators such as concentrations of PTEs with low coefficient of variation are generally enunciated as natural resources while the higher coefficient of variation is typically expressed by manmade pollution (Baltas et al., 2020; Cai et al., 2015; Jiang et al., 2019). The coefficients of variation for As, Cr, Cu, Ni, Pb, and Zn were 20.19%, 32.06%, 29.47%, 21.69%, 47.60%, and 10.68%, respectively (Table 1). The coefficients of variation values of six PTEs contents in the study region soils followed a descending order as: Pb>Cr>Cu>Ni>As>Zn (Table 1). The coefficients of variation for Zn was very smaller than those of the other PTEs in the study region, indicating that Zn has a weak variability (CV<25%). It is assumed that the inputs of this metal in the study region may be controlled by the patent material of the soil and also topography. The coefficients of variation of Pb was the highest of all studied PTEs, signifying that Pb has the largest variation among the soil samples in the study region. Additionally, coefficients of variation for As, Cr, Cu, Ni and Pb were larger than 20% but lower than 50%, demonstrating the moderate degree of variations in the soils of the investigated region. The fluctuations in the coefficients of variation could be due to the discrete inputs related to natural or external factors (Adimalla et al., 2020; Jiang et al., 2019).

3.2 Heavy metals spatial distribution

The Spatial distribution patterns of six priority PTEs measured in the surface soils of the study region were depicted in Fig 2. As shown in Fig 2, the spatial distribution patterns of As and Pb established a quite similar trend that their contents were higher in the northwestern and southeastern directions of *Malkalapad* town/city. The higher concentration of Zn was found in 60% of the study region and mainly in the southern region as the site is adjacent to the main highway with numerous roads, transportation hubs with bus stations. Consequently, vehicle exhaust seems to be a noticeable source of pollution towards Zn. The spatial distribution of Ni exhibited the higher concentration of Ni was measured at *ZSI-10* (51 mg/kg) in the proximity to the *Bardipur* town which is located in the southern part of *Malkalapad* city (Fig 2). This could be due to parent rock materials or atmospheric deposition of vehicle emissions (Huang et al., 2019; Wang et al., 2019; Zhao et al., 2019). However, concentrations of Ni decreased in the vicinity of *Kottur* and the northeastern part of the study region. The entire study region has a very high Cr and Cu concentrations, basically 6.03 and 3.45 times higher than their geochemical background values (Fig 2). The spatial distribution pattern of Cr and Cu was similar, and very high pollution was noticed in the vicinity of the western part of the investigated region. It is noted that Cr and Cu metals had higher skewness and their contribution is also quite higher in the risk screening in the study region.

3.3 Pollution assessment of heavy metals

3.3.1 Contamination factor (*CF*) and degree of contamination (*DC*)

In order to evaluate the level of contamination and possible anthropogenic inputs in the soil samples, the contamination factor (*CF*) and degree of contamination (*DC*) were computed for selected six PTEs in the present study. The computed *CF* and *DC* values for six PTEs are listed in Table 2. The mean *CF* values of the six PTEs in this study follow a descending order as Zn (9.01)>Cr (6.60)>As (2.29)>Pb (1.47)>Cu (1.22) >Ni (1.10). The *CF* ranges of As, Cr, Cu, Ni, Pb, and Zn are 1.44-3.00, 4.94-15.06, 0.85-2.16, 0.64-1.41, 0.27-2.81, and 7.37-11.75, respectively. And classification of mean *CF* is also depicted in Fig 3. As shown in Fig 3, the average *CF* value for As, Cu, Ni, and Pb showed a moderate contamination level, whereas the mean *CF* values for both Zn and Cr in the soils showed as very high contamination levels which indicates that the soil of the present study is considered to very highly polluted (Fig 3). Based on the CF values, and degree of contamination (*DC*) values are generally computed to systematically assess the soil pollution statuses in the investigated region. Therefore, the *DC* values ranged from 15.50 to 36.50 with a mean of 21.69 (Table 2), indicating the soil sites are polluted by a moderate degree of contamination to very high degree of contaminated could be due to the influence of external discrete sources such as human activities and other anthropogenic inputs (Ali et al., 2019; Jiang et al., 2019).

3.3.2 Evaluation of Index of geo-accumulation

The index of geo-accumulation (I_{geo}) is mostly used model to assess the cumulative pollution level for PTEs in soils all over the world (Kumar et al., 2019; Muller, 1969; Pobi et al., 2020; Said et al., 2019). The extent of PTEs pollution in soils of the investigated region was evaluated using the index of geo-accumulation and obtained results were shown in Table 2. Moreover, the distribution map of I_{geo} for six PTEs is depicted in Fig 3. The range of I_{geo} values for the studied six PTEs i.e., As, Cr, Cu, Ni, Pb and Zn were were -0.06-1.00±0.58, 1.72-3.33±2.09, 1.16-2.51±1.63, -1.23-0.19±0.48, -2.46-0.90±-0.25, and -0.35-0.33±-0.07, respectively (Table 2 & Fig 3). It can be obviously seen from the Table 2, the I_{geo} values for Ni, Pb and Zn were smaller than 1 at all the soil sampling sites, signifying that soil of the study region was viewed as uncontaminated to moderately contaminated by metals of Ni, Pb and Zn. The I_{geo} for Cr at site ZSI-4 showed the highest value reached 3.33 and remaining soil sampling sites were lower than 3, indicating that the soils of the investigated region were moderate to heavily contaminated by chromium. Meanwhile, the I_{geo} for Cu at sites ZSI-5, ZSI-6, and ZSI-19 signifying moderately to heavily contaminated and remaining sites were moderately contaminated. The I_{geo} values for As in most of the sampling sites were lower than zero, thus those sampling sites in the study region were noticed as not polluted.

3.3.3 Principal component analysis (PCA) for heavy metals in soil

In this study, we applied the varimax rotation-Kaiser Normalization method, in order to obtain the principal component analysis (PCA) for six PTE concentrations in soils and results are listed in Table 3. As can be seen from Table 3, two principal components with eigenvalues larger than unity (1.0) were obtained, which typically elucidated nearly 58% of the data variability. The first principal component (PC1) which essentially contained As (0.849) and Pb (0.925) loads were very high, contributing to 39.999% of the total variance and also showed an eigenvalue of 2.24 (Table 3). The second principal component (PC2) accounts for over 17% of the total variance, and showing weak positive loading for Zn (0.481) and Ni (0.346) and remaining PTEs loads are quite low. This could be due to that they have some inimitable source by both anthropogenic and natural activities. Furthermore, it is observed that the mean concentrations of As, Zn and Ni were very larger than their corresponding geochemical background values which indicating that these three PTEs are typically from geochemical weathering of parent rock material. The researchers of Adimalla et al. (2020), Jiang et al. (2019) and Chen et al. (2016) have also identified that the road and population densities, vehicle exhaust emissions, tire wear, land use types, especially weathering of host rocks, intensive human activities, and improper disposal of domestic wastes are the most significant indicators of heavy metals to accumulate in the urban soils.

3.4 Potential human health risk assessment

According to the method of human health risk assessment suggested by the USEPA, the non-carcinogenic and carcinogenic health risk of soil PTEs can be assessed and computed based on three potential routes including ingestion, inhalation and dermal contact. The obtained results are listed in Table 4. It is evidently observed from Table 4, the values of *HQ* and *CR* followed the decreasing order of exposure pathways: ingestion>dermal>inhalation for both adults and children in the study region. This finding obviously suggests that the ingestion of soil PTEs is the principal key factor that is most likely to impact on health risks in the surveyed region. However, in this study, $HQ_{ingestion}$, $HQ_{inhalation}$, and HQ_{dermal} values of six PTEs for adults were marginally lower than those for children in the study region (Table 4). In other words, children in the study region have greater non-carcinogenic risk than adults through all three exposure pathways which are described above. Recent studies have also discovered that higher soil ingestion and lower body weight are the two major causes of health risks in children (Adimalla et al., 2020; Chen et al., 2016; Jiang et al., 2020). For the ingestion exposure pathway for adults and children, the non-carcinogenic risk decreased as follows: Cr>As>Pb>Ni>Cu>Zn, suggesting the contribution of Cr in non-carcinogenic risk is greater than other five PTEs. It was observed from Table 4, that non-carcinogenic risk (*HI*) values of Cr, Cu, Zn, Pb, Ni, and As for adults were varied from 9.10E-02 to 3.44E-04, 3.04E-03 to 7.75E-03, 4.08E-04 to 6.51E-04, 1.30E-03 to 1.34E-02, 1.38E-03 to

3.70E-03, and 1.10E-02 to 2.30E-02, while children were 6.01E-01 to 1.83E+00, 2.12E-02 to 5.40E-02, 2.84E-03 to 4.53E-03, 9.02E-03 to 9.31E-02, 9.60E-03 to 2.58E-02, and 7.69E-02 to 1.60E-01, respectively. Results indicate that for children and adults, except metal Cr, the *HI* seemed to be lower than unity, indicating have no serious health risk for both age groups (children and adults) in the study region. Predominantly, for children, the *HI* values of Cr were very higher than unity (*HI*>1), this situation demonstrates that children are more sensitive to the adverse health effects of PTEs in the investigated region (USEPA, 1989, 1997).

Due to the lack of the carcinogenic slope factors for Cu, Ni and Zn, only the carcinogenic risks for the other three PTEs (As, Cr and Pb) were computed in the study region, and also results were listed in Table 4. The value of total carcinogenic risk (*TCR*) ranges from 3.78E-08 to 3.46E-04 with a mean of 7.91E-05 for adults, while the *TCR* values for children range from 2.64E-07 to 2.42E-03 with a mean of 5.53E-04. For children and adults, the carcinogenic risk caused by Cr is greater than that of As and Pb. The calculated *TCR* values varied as Cr>As>Pb for children and adults in the study region. As Table 4 shows, Cr accounts for the majority of carcinogenic health risks for especially children. The *TCR* of Cr, As, Pb was all lower than the recommended limit of 1.00E-04 for adults, while the *TCR* for children was 5.53 times higher than the acceptable limit. This finding shows that children in the study region typically constitute a major health risk. However, adults have no effective health risks due to *TCR* values are quite lower than the recommended limit (Table 4). Overall, health risk assessment suggesting the necessary precautions should be taken in order to protect the children's health and also reduce the impact of health risk in the study region.

4 Conclusions

In this study we used contamination factor, degree of contamination, index of geo-accumulation and principal component analysis to explore the contamination status by PTEs (As, Cr, Cu, Ni, Pb, and Zn) and also we evaluated human health risk to children and adults in the urban region of south India. The results show that Cr, Cu, Ni and Zn contents were 6.03, 3.45, 1.64, and 1.45 times greater than their corresponding geochemical background values, respectively. The results of a series of model estimation indices including *CF*, *DC*, and I_{geo} suggest that soil of the investigated region is majorly moderate contamination to high contamination due to various discrete sources. The soil PTEs typically pose both non-carcinogenic and carcinogenic risks to the children and adults health risks predominantly through Cr and As emissions. The main exposure pathway was identified as ingestion for both non-carcinogenic and carcinogenic risks in the study region. However, non-carcinogenic risks for children and adults in the examined region were within the secure limits, indicating no non-carcinogenic risk, while carcinogenic risk has a significant risk to the children in the study region. Therefore, necessary precautionary measures can be implemented in order to reduce the health risks in the study region.

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Table 1. Descriptive statistics for PETs (mg/kg) in soils from the study region.

Heavy metals	As	Cr	Cu	Ni	Pb	Zn
Minimum	2.3	158	84	19	3.1	84
Maximum	4.8	482	214	51	32	134
Mean	3.665	211.165	120.6	32.8	16.715	102.75
Median	3.65	198	112	31.5	17	103.5
25^{th} Percentiles	3.1	185.15	98	28	10.35	93.5
75^{th} Percentiles	4.2	210.5	133	37	23	107
Standard deviation	0.74	67.70	35.54	7.11	7.96	10.97
CV%	20.19	32.06	29.47	21.69	47.60	10.68
Skew	0.057	3.662	1.483	0.556	-0.006	0.931
Kurtosis	-0.847	15.052	1.866	1.158	-0.637	2.364
K-S	0.089	0.296	0.218	0.1	0.07	0.166

K-S: Kolmogorov-Smirnov statics'
CV%: Coefficient of variation

Table 2. Contamination factor (*CF*) and degree of contamination (*DC*) for six PETs in the study region soils

	Contamination factor (*CF*)			Index of geo-accumulation (I_{geo})		
Metals	minimum	maximum	mean	minimum	maximum	mean
As	1.44	3.00	2.29	-0.06	1.00	0.58
Cr	4.94	15.06	6.60	1.72	3.33	2.09
Cu	0.85	2.16	1.22	1.16	2.51	1.63
Ni	0.64	1.71	1.10	-1.23	0.19	-0.48
Pb	0.27	2.81	1.47	-2.46	0.90	-0.25
Zn	7.37	11.75	9.01	-0.35	0.33	-0.07
Degree of contamination (*DC*)	15.50	36.50	21.69	/	/	/

Table 3. Total variance explained and matrix of principal components analysis

Total Variance Explained	Initial Eigenvalues			Component		
	Total	% of Variance	Cumulative %	PETs	PC1	PC2
1	2.400	39.999	39.999	As	**0.849**	0.279
2	1.064	17.733	57.732	Cr	0.200	-0.708
3	0.949	15.824	73.556	Cu	-0.652	0.287
4	0.887	14.791	88.347	Ni	0.345	0.346
5	0.647	10.775	99.123	Pb	**0.925**	0.225
6	0.053	0.877	100.000	Zn	-0.490	0.481

Extraction method: principal component analysis. Rotation method: Varimax with Kaiser Normalization; PC1 is the first principal component, PC2 is the second principal component, significant loading factors are remarked in bold

Table 4. The results of health risk assessment (non-carcinogenic and carcinogenic risks) of soil heavy metals from different sources

			Non-carcinogenic risks				Carcinogenic risks			
PETs	Groups		HQ_{ing}	HQ_{inh}	HQ_{dermal}	HI	CR_{ing}	CR_{inh}	CR_{dermal}	TCR
Cr	Adult	Minimum	7.52E-02	7.43E-04	1.50E-02	9.10E-02	1.13E-04	1.06E-08	4.50E-07	1.13E-04
		Maximum	2.30E-01	2.27E-03	4.58E-02	2.78E-01	3.44E-04	3.24E-08	1.37E-06	3.46E-04
		Mean	1.01E-01	9.93E-04	2.01E-02	1.22E-01	1.51E-04	1.42E-08	6.02E-07	1.51E-04
	Children	Minimum	5.27E-01	4.43E-04	7.37E-02	6.01E-01	7.90E-04	6.33E-09	2.21E-06	7.92E-04
		Maximum	1.61E+00	1.35E-03	2.25E-01	1.83E+00	2.41E-03	1.93E-08	6.75E-06	2.42E-03
		Mean	7.04E-01	5.92E-04	9.85E-02	8.03E-01	1.06E-03	8.46E-09	2.96E-06	1.06E-03
Cu	Adult	Minimum	3.00E-03	2.82E-07	3.99E-05	3.04E-03	/	/	/	/
		Maximum	7.64E-03	7.19E-07	1.02E-04	7.75E-03	/	/	/	/
		Mean	4.31E-03	4.05E-07	5.73E-05	4.36E-03	/	/	/	/
	Children	Minimum	2.10E-02	1.68E-07	1.96E-04	2.12E-02	/	/	/	/
		Maximum	5.35E-02	4.29E-07	4.99E-04	5.40E-02	/	/	/	/
		Mean	3.02E-02	2.42E-07	2.81E-04	3.04E-02	/	/	/	/
Zn	Adult	Minimum	4.00E-04	3.76E-08	7.98E-06	4.08E-04	/	/	/	/
		Maximum	6.38E-04	6.01E-08	1.27E-05	6.51E-04	/	/	/	/
		Mean	4.89E-04	4.61E-08	9.76E-06	4.99E-04	/	/	/	/
	Children	Minimum	2.80E-03	2.24E-08	3.92E-05	2.84E-03	/	/	/	/
		Maximum	4.47E-03	3.58E-08	6.25E-05	4.53E-03	/	/	/	/
		Mean	3.43E-03	2.75E-08	4.80E-05	3.47E-03	/	/	/	/
Pb	Adult	Minimum	1.27E-03	1.18E-07	3.37E-05	1.30E-03	3.76E-08	3.54E-12	1.97E-08	3.78E-08
		Maximum	1.31E-02	1.22E-06	3.47E-04	1.34E-02	3.89E-07	3.66E-11	4.10E-08	3.90E-07
		Mean	6.82E-03	6.38E-07	1.81E-04	7.00E-03	2.03E-07	1.91E-11	3.13E-08	2.04E-07
	Children	Minimum	8.86E-03	7.06E-08	1.65E-04	9.02E-03	2.64E-07	2.11E-12	9.66E-08	2.64E-07
		Maximum	9.14E-02	7.29E-07	1.71E-03	9.31E-02	2.72E-06	2.18E-11	2.02E-07	2.73E-06
		Mean	4.78E-02	3.81E-07	8.91E-04	4.86E-02	1.42E-06	1.14E-11	1.54E-07	1.42E-06
Ni	Adult	Minimum	1.36E-03	1.24E-07	2.01E-05	1.38E-03	/	/	/	/
		Maximum	3.64E-03	3.33E-07	5.38E-05	3.70E-03	/	/	/	/
		Mean	2.34E-03	2.14E-07	3.46E-05	2.38E-03	/	/	/	/

	Children	Minimum	9.50E-03	7.39E-08	9.85E-05	9.60E-03	/	/	/	/
		Maximum	2.55E-02	1.98E-07	2.64E-04	2.58E-02	/	/	/	/
		Mean	1.64E-02	1.28E-07	1.70E-04	1.66E-02	/	/	/	/
As	Adult	Minimum	1.10E-02	2.51E-06	4.37E-05	1.10E-02	4.93E-06	4.64E-10	1.50E-10	4.95E-06
		Maximum	2.29E-02	5.25E-06	9.12E-05	2.30E-02	1.03E-05	9.68E-10	1.55E-09	1.03E-05
		Mean	1.75E-02	4.01E-06	6.96E-05	1.75E-02	7.85E-06	7.39E-10	8.10E-10	7.89E-06
	Children	Minimum	7.67E-02	1.50E-06	2.15E-04	7.69E-02	3.45E-05	2.77E-10	7.38E-10	3.46E-05
		Maximum	1.60E-01	3.13E-06	4.48E-04	1.60E-01	7.20E-05	5.77E-10	7.62E-09	7.22E-05
		Mean	1.22E-01	2.39E-06	3.42E-04	1.23E-01	5.50E-05	4.41E-10	3.98E-09	5.51E-05

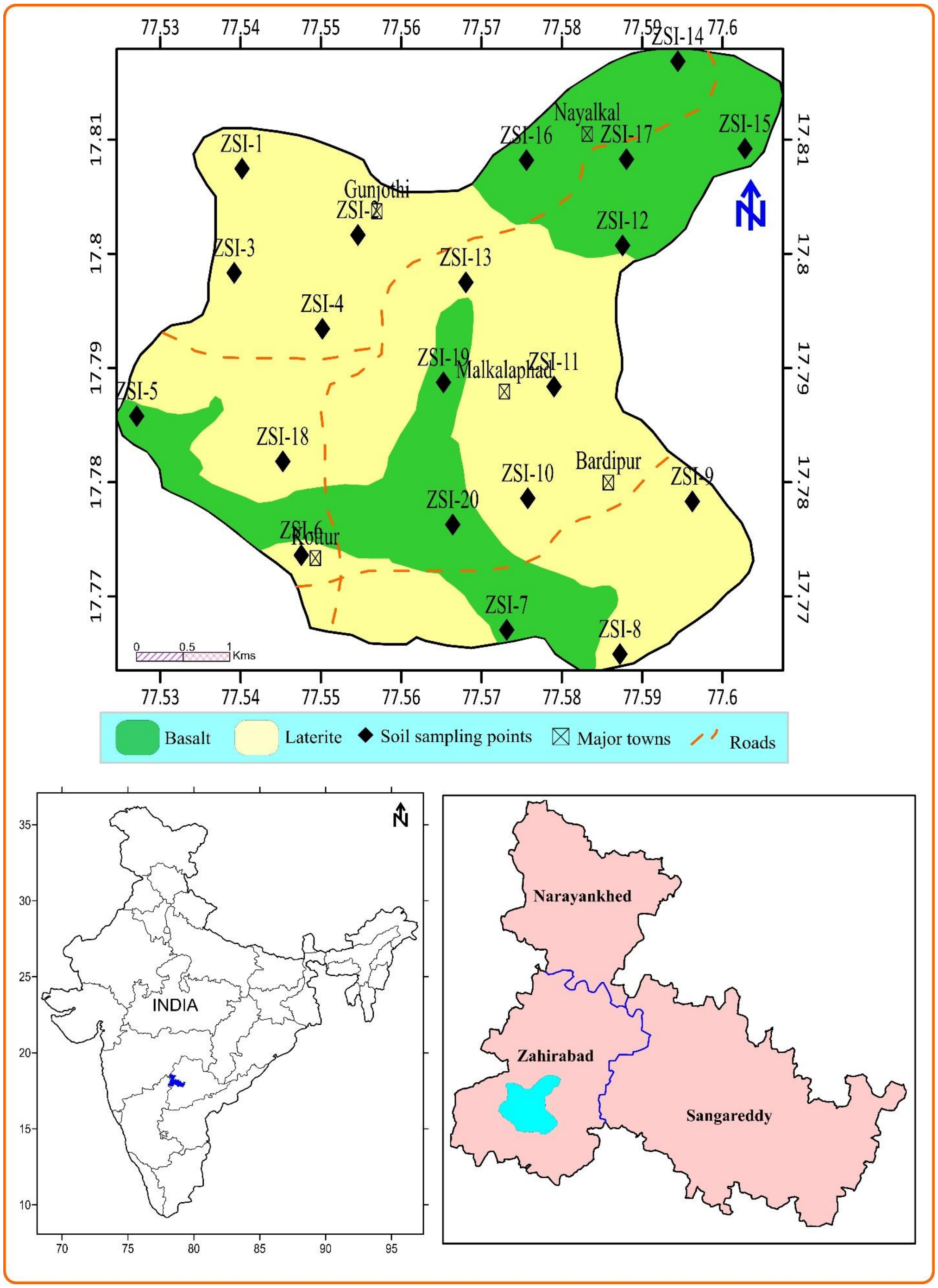

Figure 1. Location map of the examined region showing soil sampling sites, major residential/towns, major roads and geological pattern of the study region. Samples ZSI-5, ZSI-7, ZSI-12, ZSI-14, ZSI-15, ZSI-16, ZSI-17, ZSAI-19, and ZSI-20 were collected in Basalt region, and remaining samples ZSI-1 to ZSI-4, ZSI-6, ZSI-8 to ZSI-11, ZSI-13 and ZSI-18 were located in the laterite region of the study region.

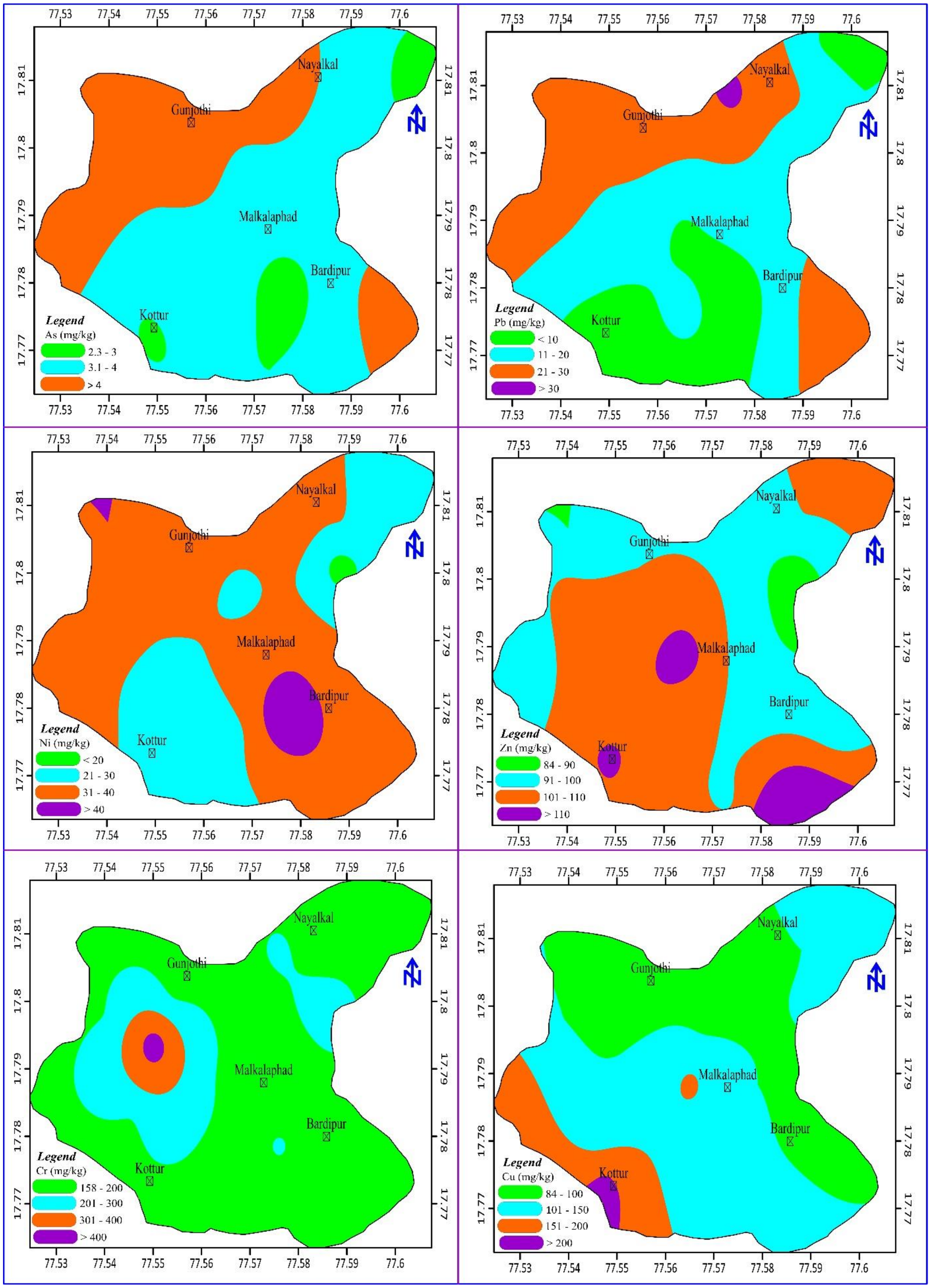

Figure 2. Spatial distribution patterns of potentially toxic elements (PTEs) (Arsenic, lead, nickel, zinc, chromium and copper) in the soils of the south India.

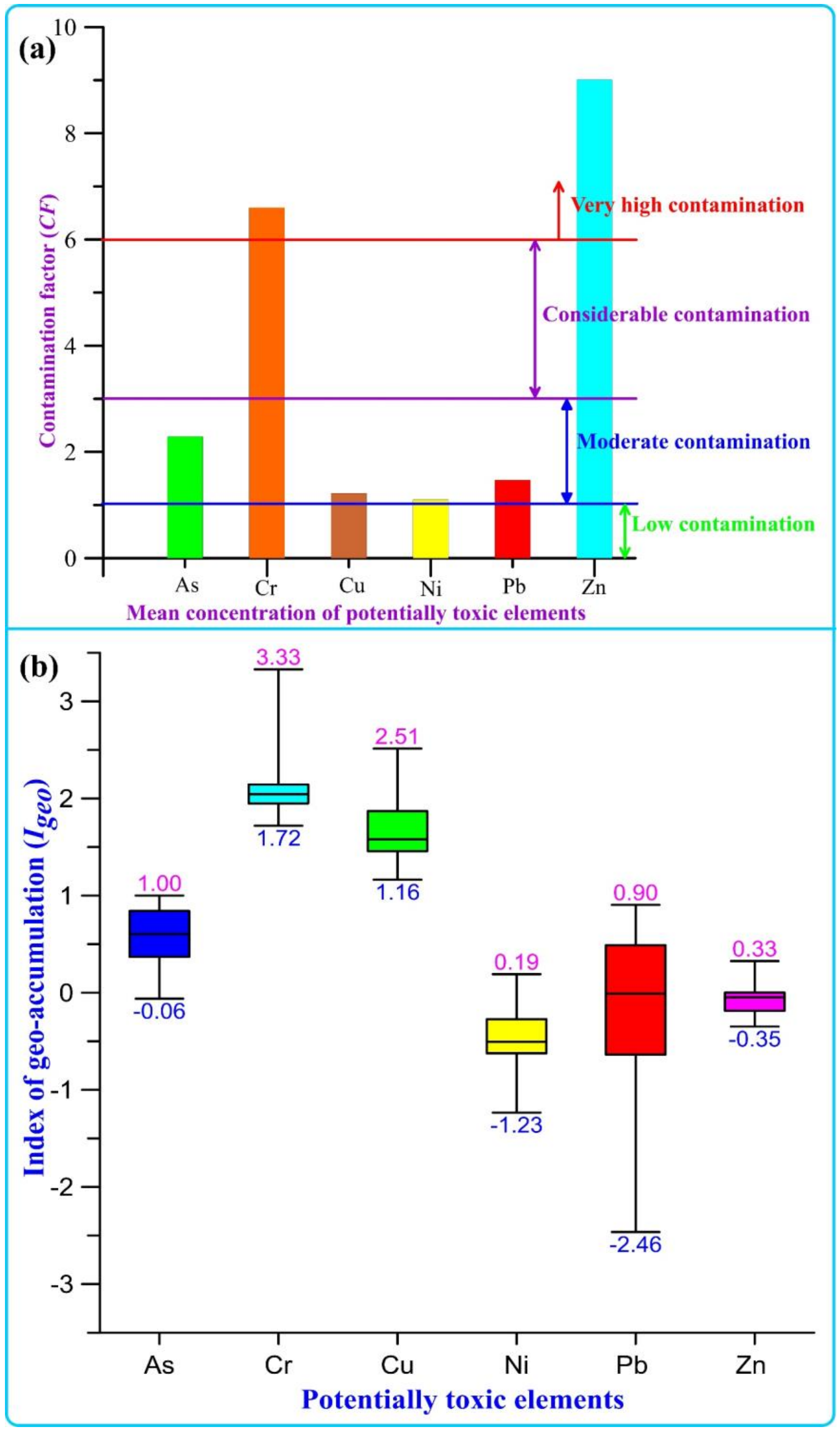

Figure 3. (a) The mean values of contamination factor (*CF*) of six potentially toxic elements (PTEs) in soils of urban region of south India (Green 2-stick heads represents the low contamination factor (*CF*<1); blue one signifies the moderate contamination (1≤*CF*≤3); purple one denotes the considerable contamination (3≤*CF*≤6) and red one symbolizes the very high contamination (*CF*>6). (b) The index of geo-accumulation (I_{geo}) of six heavy metals in the soils of the study region.

SUPPLEMENTARY DATA

Potentially toxic elements (PTEs) pollution in surface soils in a typical urban region of south India: An application of health risk assessment and distribution pattern

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Table S1. Results of analytical values*** of the standard soil reference materials SO-1 (regosolic clay soil) and SO-4 (chermozemic A horizon soil) in comparison with the certified reference values

CRM	As	Cr	Cu	Pb	Ni	Zn
*SO-1	2	170	61	20	92	140
**Tested values	1.96	167.2	60.4	19.5	91.6	138.6
% of accuracy	98.00	98.35	99.02	97.50	99.57	99.00
*SO-4	7.4	64	21	14	24	94
**Tested values	7.19	63.5	20.8	12.9	23.5	93.1
% of accuracy	97.16	99.22	99.05	92.14	97.92	99.04

* suggest the certified values

** indicate the measured/tested values (n = 3)

***The recovery rates of the target PTEs in the standard references ranged from 97.5% to 99.02%.

Table S2. Parameters used for calculation of the average daily exposure to potentially toxic elements (PTEs)

Items	Parameters	Meaning	Unit	Value for children	Value for adults
Basic parameters	C_{soils}	Heavy metal concentrations	mg/kg	Present study results	Present study results
Exposure behavioral parameters	*EF*	Exposure frequency	days/year	350	350
	ED	Years of exposure	years	6	24
	BW_A	Average body weight	Kg	15	55.9
	ET_A	Average exposure time	days	365×ED (Non-carcinogenic effect)	365×ED (Non-carcinogenic effect)
				365 × 70 (Carcinogenic effect)	365 × 70 (Carcinogenic effect)
Hand–mouth intake	*IngR*	Ingestion rate of soil	mg/day	200	100
Respiratory intake	*InhR*	Inhalation rate of soil	m^3/day	7.6	20
Skin contact	ESA_S	Exposed skin surface area	cm^2	2800	5700
	AF_S	Soil to skin adherence factor	mg/cm^2	0.2	0.07
	EF_p	Particle emission factor	m^3/kg	1.36×10^9	1.36×10^9

Source: (Adimalla et al., 2020; Baltas et al., 2020; USEPA, 1989, 1997, 2002)

Table S3. Values of reference doses (*RfD*: mg/kg/day) and slope factors (SF: per mg/kg/day) for five PETs

Exposure pathway		Cr	Pb	Cu	Zn	Ni
RfD	Ingestion	3.00E-03	3.50E-03	4.00E-02	3.00E-01	2.00E-02
	Dermal absorption	6.00E-05	5.25E-04	1.20E-02	6.00E-02	5.40E-03
	Inhalation	2.86E-05	/	/	/	9.00E-05
SF	Ingestion	5.00E-01	8.50E-03	/	/	/
	Dermal absorption	/	/	/	/	/
	Inhalation	4.20E+01	/	/	/	8.40E-01

Definitions and reference values of both non-carcinogenic and carcinogenic risks presented in equations 5 to 12 are clearly recorded in Table S2 as obtained from the relevant literature (Adimalla et al., 2020; Baltas et al., 2020; USEPA, 1989, 1997, 2002). Similarly, reference dose and slope factors values are also very important in order to assess the health risk assessment in the study region. Without Table S2 & S3 values it is very difficult to compute the non-carcinogenic and carcinogenic risks in any region. Therefore, we used above parameters and its values to evaluate the health risk for children and adults in the study region.

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